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SUBJECT: AN ANALYSIS OF THE POTENTIAL HAZARDS ASSOCIATED WITH THE
DISPOSAL OF RADIOACTIVE WASTE IN OPEN PITS AT ORNL

TO: Distribution

FROM: F. T. Binford

An examination of the present procedure for the disposal of radioactive waste in open pits at ORNL discloses that the activity discharged to the environment from this source has in the past represented a small fraction of the total contribution to the MPC ratio in the river. It appears, moreover, that, if certain limitations are observed, this method of handling waste is more satisfactory from a hazard standpoint than others in current use.

Several areas where additional information is needed have become apparent, and it is recommended that steps be taken to rectify this situation.

This document has been approved for release
to the public by:

David E. Hamlin 3/4/96
Technical Information Officer Date
ORNL Site

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AN ANALYSIS OF THE POTENTIAL HAZARDS ASSOCIATED WITH
THE DISPOSAL OF RADIOACTIVE WASTE IN
OPEN PITS AT ORNL

F. T. Binford

INTRODUCTION

This study was undertaken at the request of the Waste Effluents Committee in order to place in its proper perspective the hazard which arises from the use of pits for the disposal of radioactive waste at ORNL. Much of the information contained herein was obtained with the aid and cooperation of the members of the Health Physics Division. The assistance of E. G. Struxness, H. H. Abee, and K. E. Cowser of that Division was in a large measure responsible for the compilation of the information contained herein. Supporting information for many of the statements is contained in CF-60-5-29 edited by E. G. Struxness.

METHOD OF PROCEDURE

The hazards associated with waste pits used for the disposal of liquid waste at ORNL can be conveniently divided into two categories as follows:

(1) Hazards which result because of continual seepage of radioactive material from the pits and the subsequent introduction into the river of some fraction of this material.

(2) Hazards which exist because of the presence in and near the pits of a sizeable inventory of radionuclides with the accompanying possibility that a significant fraction of this inventory could be introduced into the river in an uncontrolled fashion. This category can be further subdivided into two general classes of hazards: First, those

associated with the presence of radioactive material contained in solution or in suspension in the pits themselves and second, those associated with the inventory of radionuclides fixed or semifixed by sorption in the soil near the pits.

In order to estimate the magnitude of these hazards, it is first necessary to establish the rate at which mixtures of radioisotopes may be safely discharged to the river. This is accomplished through the use of current MPC values as set forth in Bureau of Standards Handbook 69⁽¹⁾ together with a knowledge of the flow in the river. Once these numbers are known, the various sources of contamination must be evaluated in order to determine their contribution to the total concentration in the river. Next, general criteria are established for the regulation of these sources. These criteria are designed to insure that the sources, and in particular the waste pits, are handled normally in such a way that the maximum safe concentrations are not exceeded.

It is at once obvious that the criteria assigned for the regulation of the waste pit operation must depend strongly upon the behavior of the other sources of contamination. It is, in general, not possible to treat any one waste disposal operation as independent of the others, but each must be considered in relation to all of the others.

Finally, an attempt is made to assess the likelihood and the consequences of abnormal conditions which could result in a hazardous situation. To be realistic, it is here necessary to examine the "incremental hazard." By this is meant the difference between the over-all hazard

associated with the waste disposal system with the waste pits in service and the same hazard with the waste pits out of service.

In making this analysis, it has been tacitly assumed that the river itself is an ultimate means of waste disposal and that it is the intention to discharge into it as much radioactive material as it will safely take. It follows that the reduction of the rate at which contamination is discharged to the river to a value very much less than that established as a safe upper limit is not a desirable goal in itself. On the contrary, there is an advantage in discharging to the river as much radioactivity as possible without exceeding the safe limits. It is clearly also advantageous to decrease the amount of radioactivity discharged to the river per unit processed in the primary waste disposal operations in order to increase the capacity of these primary operations. This philosophy, which may at first seem startling, can be readily justified on an operational basis provided that the word "safe" in the foregoing context really carries the usual dictionary meaning "secure from threat of harm." In the sequel it will be assumed that the concentration standards are so chosen that, when we say "safe", we really mean "safe." It is recognized that in its present state our knowledge of the hazards associated with the release of radioactive material to the environment is inadequate to provide a reliable value for this quantity. It is necessary, therefore, to select a reasonable number based upon the best available information and to admit the likelihood that it will be revised if and when better information is forthcoming. Once this number is chosen, it is possible to formulate

operating criteria in terms of a definite upper limit rather than in terms of an indefinite lower limit.

THE ESTABLISHMENT OF ALLOWABLE DISCHARGE RATES

The beta-emitting radioelements which have been found in significant amounts in the waste reaching the Clinch River from the Laboratory are listed for the period 1953-1959 in Table I. In this table, it should be noted that the column headed TRE consists of a mixture of the trivalent rare earths, primarily Y^{90} , in equilibrium with Sr^{90} . Since the presence of Y^{90} is already considered in the calculation of the MPC for Sr^{90} , only the difference between the TRE fraction and the strontium fraction will be considered. When this difference is positive, it will be taken to be Y^{91} ; when it is negative, it will be neglected.

Corresponding analyses for the quantity of α emitters discharged to the river are not available; however, the totals have been estimated and are listed in Table II.

TABLE II⁽³⁾

Yearly Discharge of α Emitters into the Clinch River
(Computed as Curies of Pu^{239})

<u>Year</u>	<u>Curies</u>
1959	0.68*
1958	0.08
1957	0.15
1956	0.28
1955	0.25
1954	0.07
1953	0.08

*Probably due to 3019 leak.

TABLE I (2)

Yearly Discharges of Radioactivity to Clinch River
1953 - 1959

Year	Gross Beta ^a Curies	TRE ^c (CO)	Per Cent of Gross Beta Activity Identified With Specific Radionuclides ^b								
			Ce	Ru	Zr	Cs	I	Sr	Nb	Ba	Co
1953	304	37.4	2.2	8.7	2.5	2.1	0.7	44.3	1.2	0.9	---
1954	384	42.6	6.2	2.9	3.6	5.7	0.9	35.2	2.4	0.6	---
1955	437	33.7	19.4	7.0	1.2	14.4	1.6	21.1	1.3	0.2	1.5
1956	582	23.8	10.1	5.0	2.0	29.6	0.6	17.9	2.6	0.5	7.9
1957	397	28.8	3.2	15.0	5.7	22.4	0.3	20.9	1.8	0.4	1.2
1958	544	43.8	5.5	7.7	1.1	10.1	1.5	27.0	1.1	0.6	1.6
1959	937	20.8	9.8	47.4	1.5	6.0	0.1	Sr ⁸⁹ 0.7 Sr ⁹⁰ 7.6	1.4	0.2	3.5

^aGross beta activity in daily samples from Creek at White Oak Dam.^bDaily samples at Dam composited for monthly radiochemical separations and analysis.^cTrivalent rare earths exclusive of cerium

The MPC values for continuous occupational use are given in Bureau of Standards Handbook 69⁽¹⁾. In order to compute the ratio of the concentration of a given mixture of radioisotopes to the MPC for that mixture, it is necessary to consider the fact that different isotopes affect different organs so that in some cases the effects of the components of the mixtures are additive. The procedure can be simplified conservatively by using the reciprocal of the harmonic sum of MPCs listed for each nuclide as the MPC to be used. For example, in the case of Sr⁹⁰ the occupational MPCs for water are given as follows: to the bone (the critical organ), 10^{-6} μ c/ml; whole body, 4×10^{-6} μ c/ml; to the GI tract, 5×10^{-4} μ c/ml. The harmonic sum is

$$\frac{1}{10^{-6}} + \frac{1}{4 \times 10^{-6}} + \frac{1}{5 \times 10^{-4}} = 1.252 \times 10^6$$

and the reciprocal of this is 8×10^{-7} which will be taken as the MPC value for Sr⁹⁰ in a mixture with other isotopes. Using this procedure, we can obtain MPCs for all the isotopes of interest. Since the non-occupational values are ten per cent of the occupational values, the results found in this way must be reduced by a factor of ten. Table III contains the values so determined.

The second row in Table III gives the equivalent of the isotope in question to a unit quantity of Sr⁹⁰-Y⁹⁰. Thus, for example, it requires 125 curies of Ru¹⁰⁶ or 16 curies of transuranic elements to produce the same effect as one curie of Sr⁹⁰-Y⁹⁰. By using these strontium ratios, it is possible to compute the ratio of a given mixture of these isotopes to the

TABLE III
Non-Occupational MPC Values for Mixtures
 $\mu\text{C}/\text{ml}$

Isotope	MPC	Sr Ratio
Co ⁶⁰	3.3×10^{-5}	0.0024
Sr ⁹⁰	8×10^{-8}	1.000
Y ⁹¹	3×10^{-5}	0.0027
Zr ⁹⁵	6×10^{-5}	0.0013
Nb ⁹⁵	1×10^{-4}	0.0008
Ru ¹⁰⁶	1×10^{-5}	0.008
I ¹³¹	2×10^{-6}	0.04
Cs ¹³⁷	3.9×10^{-6}	0.020
Ba ¹⁴⁰	2.5×10^{-5}	0.0032
Ce ¹⁴⁴	1×10^{-5}	0.008
U	2×10^{-5}	0.004
Trans U [*]	3×10^{-6}	0.027

*
Based on plutonium.

MPC. Letting the latter ratio be denoted by X we have

$$\frac{1}{8 \times 10^{-8}} \sum_{j=1}^{j=12} C_j r_j = X \quad (1)$$

where the C_j are the concentrations of the individual isotopes in $\mu\text{c/ml}$ and the r_j are the corresponding strontium ratios.

An examination of Tables I and II reveals that some simplifications can be made. It is clear at once that Co^{60} , Y^{91} , Zr^{95} , Nb^{95} , and Ba^{140} contribute very little to the sum. This is because these strontium ratios are low and also because they are usually present in very small amounts. Moreover I^{131} , while having a high strontium ratio, also contributes little because it is present in such minute quantities. Since uranium is probably a small contributor compared to the transuranics, we will consider all of the α emitters as plutonium. Thus we can write approximately

$$1.25 \times 10^7 \left[C_{\text{Sr}} + 0.008 (C_{\text{Ru}} + C_{\text{Ce}}) + 0.025 (C_{\text{Cs}} + C_{\alpha}) \right] \approx X \quad (2)$$

where the C's are the concentrations of the indicated nuclides, and the value 0.025 is used for the strontium ratio of both Cs^{137} and the α emitters. It is obvious that in most cases Sr^{90} will be the controlling isotope with the others contributing very little. To illustrate this, in Figure 1, C_{Sr} has been plotted versus $(C_{\text{Ru}} + C_{\text{Ce}}) + 3.125(C_{\text{Cs}} + C_{\alpha})$ for the particular cases $X = 1.0$ and $X = 0.3$. The advantage of reducing the Sr^{90} concentration below five per cent of the permitted amount is obvious from these curves.

The rate at which radioactive material can be discharged safely to the

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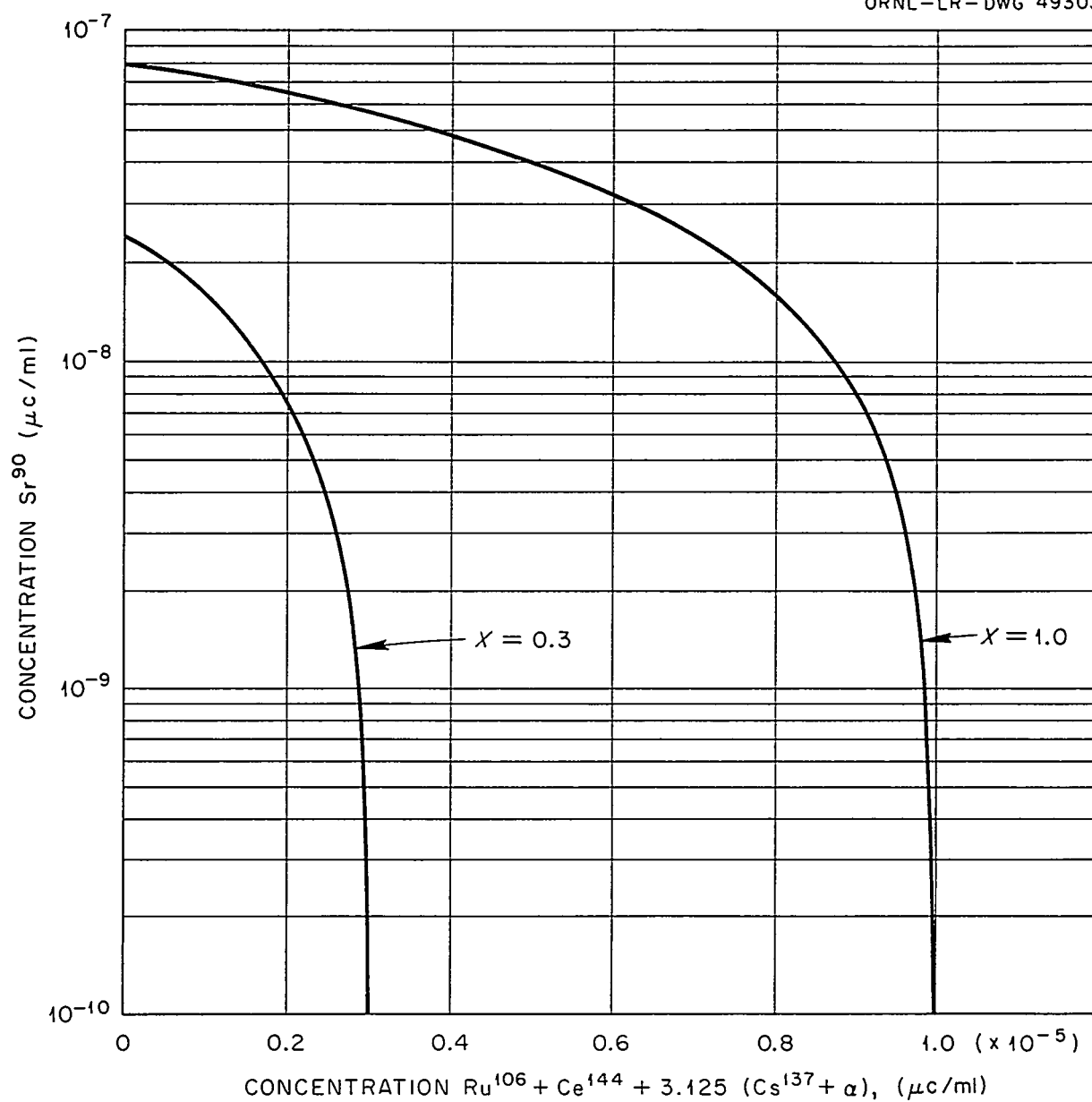


Fig. 1. Permissible Concentrations of Mixtures to Produce Various MPC Ratios.

river is governed by the requirement that the average concentration in the river shall not exceed the designated safe upper limit. It must be emphasized that this is an integral restriction. It makes little or no difference whether the limit is exceeded at times if, at other times, the concentration is sufficiently less than the limit so that the average remains below this limit. This proposition is true provided the period over which the average is taken is short compared to the life time used in arriving at the MPC values, i.e., 50 years. For operating purposes it will be convenient to choose two periods over which these averages are to be computed. These choices will be discussed later, but for the present a period of twelve months will be adopted as a standard to which the calculations can be referred.

Given a discharge rate in curies per unit time, the average concentration in the river can be computed from a knowledge of the flow rate of the river. The distribution of the weekly averaged flow rates of the Clinch River for the period 1956 through 1959 is shown in Figure 2, and the annual averages are given in Table IV.

These figures reveal that the flow in the river varies widely not only from week to week but from year to year. For simplicity we shall choose a standard flow rate; namely, 5000 CFS on which to base the calculations. The results can then be adjusted in accordance with the deviation of the actual flow from this value.

A flow rate of 5000 CFS corresponds to an annual flow of 4.46×10^9 meters³ per year. Thus, the requirement that the ratio of the average

TABLE IV
Volume Flow Rates in the Clinch River⁽⁴⁾

Year	Avg Flow (ft ³ /sec)	Total Delivered (meters ³ /yr)
1956	5150	5.59 x 10 ⁹
1957	6240	5.57 x 10 ⁹
1958	5640	5.03 x 10 ⁹
1959	3440	3.07 x 10 ⁹
Avg 4 years	5117	4.56 x 10 ⁹

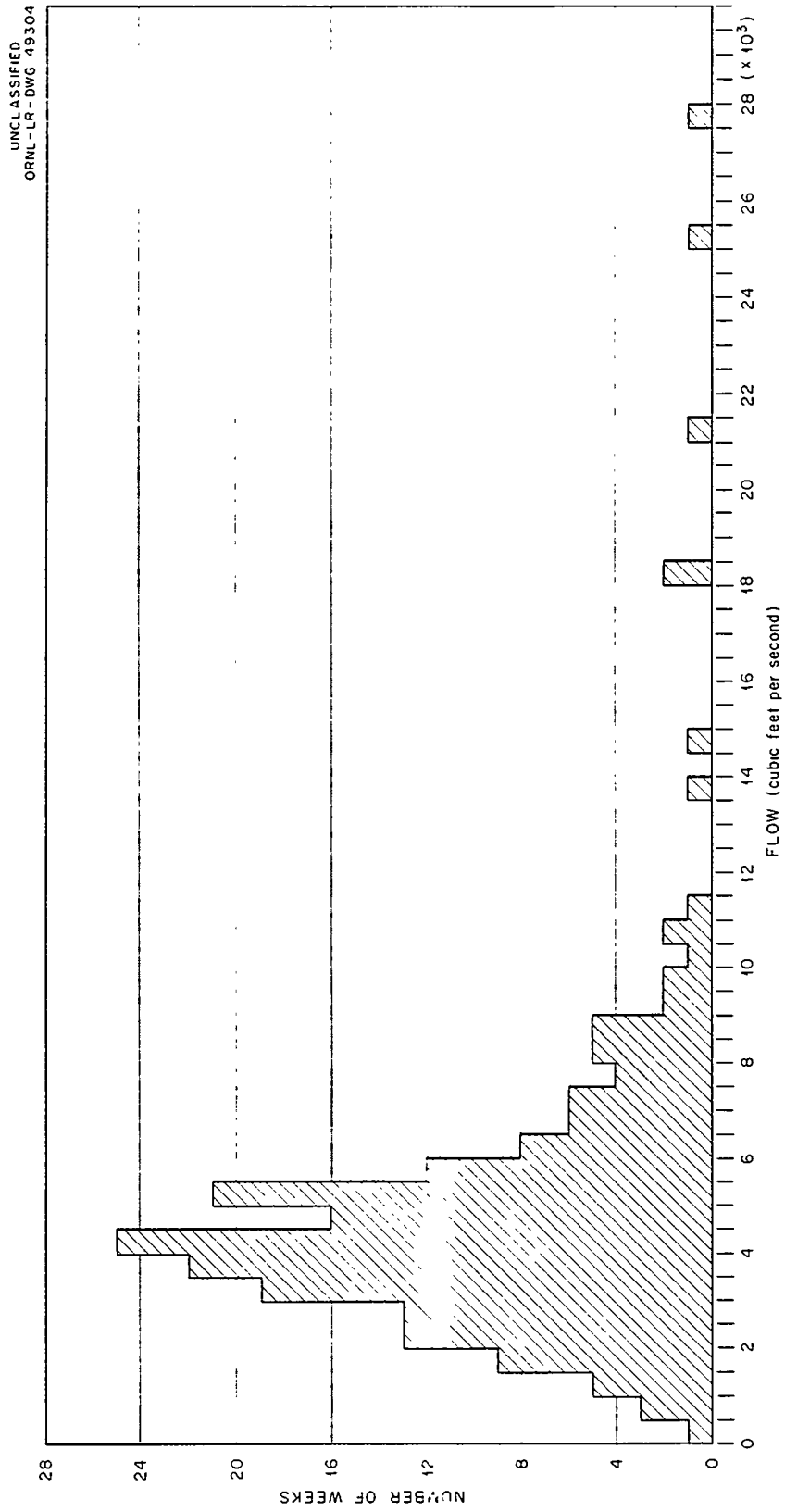


Fig. 2. Distribution of Volume Flow Rates (Weekly Averages) in the Clinch River at Mile 21 for the Period Jan 1, 1956 — Dec 31, 1959.

concentration to MPC shall be equal to or less than some value X can be expressed as

$$\frac{1.25 \times 10^7}{4.46 \times 10^9} \sum_{j=1}^{12} N_j r_j = 2.8 \times 10^{-3} \sum_{j=1}^{12} N_j r_j \leq X \quad (3)$$

where the N_j are the yearly discharges in curies to the river of the isotopes listed in Table III, and the r_j are the strontium ratios. Use of the approximation (2) yields the approximate relations

$$2.8 \times 10^{-3} \left[N_{Sr} + 0.008(N_{Ru} + N_{Ce}) + 0.025(N_{Cs} + N) \right] \leq X \quad (4)$$

Here the N's are the indicated yearly discharges in curies. Upon plotting N_{Sr} versus $(N_{Ru} + N_{Ce}) + 3.125(N_{Cs} + N)$ for the particular cases $X = 1.0$ and $X = 0.3$, the curves shown in Figure 3 are obtained. It should be recalled that these curves, as well as relations (3) and (4) are based on an average flow of 5000 CFS.

It is believed that the use of equation (4) will permit a rapid and sufficiently accurate estimate of the condition of the river for operational purposes. It cannot, however, be emphasized too strongly that this relation is valid only to the extent that the assumptions used in developing it are valid. It is intended that this relation be used to supplement, but not to replace, the present procedure of computing the result of the discharge of radioactivity based on the result of analysis of samples taken at White Oak Dam and on the measured flow in the river. It should be noted, however, that, given the analysis, a procedure similar to that used in developing

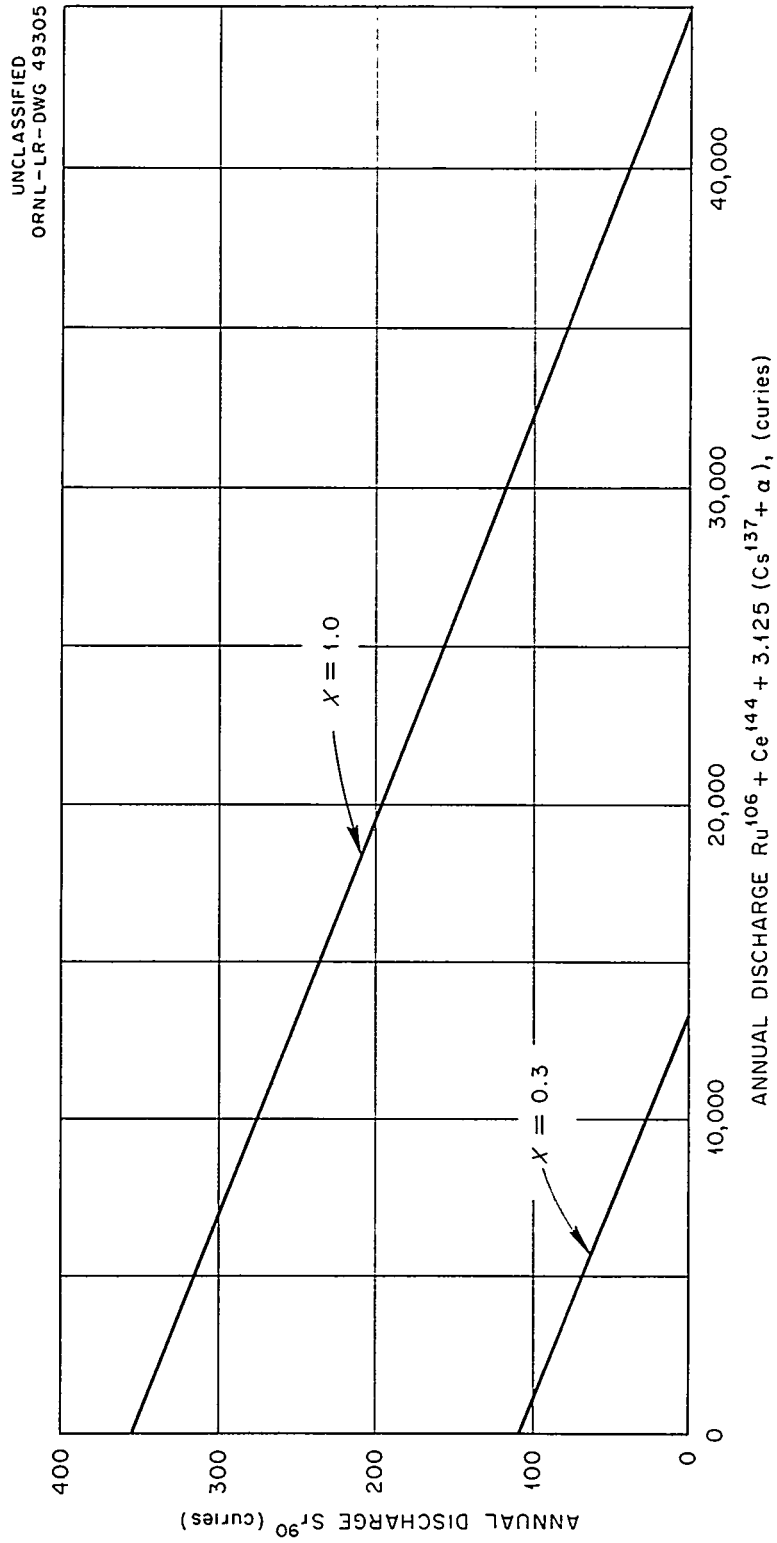


Fig. 3. Permissible Annual Discharges of Sr^{90} vs Ce^{144} , Ru^{144} , Cs^{137} and α Emitters; Based on 5000 cfs Flow.

formula (4) can be applied to the mixture so found. To illustrate the use of the formula, the ratios, X, found by this method are given in Table V for the years 1956-1959 together with those reported by the usual procedure.

TABLE V

<u>Year</u>	<u>X (From Equ 4)</u>	<u>X (Conventional Method)⁽²⁾</u>
1956	0.31	0.24
1957	0.19	0.17
1958	0.37	0.28
1959	0.34	0.26

SOURCES OF CONTAMINATION AT THE X-10 AREA

The locations of the various sources of radioactive material which may contribute to contamination in White Oak Creek and thus to the river are shown in Figure 4. These sources are listed in Table VI below. The starred items represent those which are considered to be the major sources.

TABLE VI

Sources of Radioactive Material in the X-10 Area

(A) Sources in the plant area

- Process waste system*
- Settling basin*
- Reactor ponds
- Storm sewer system
- Sewage disposal plant
- Laundry

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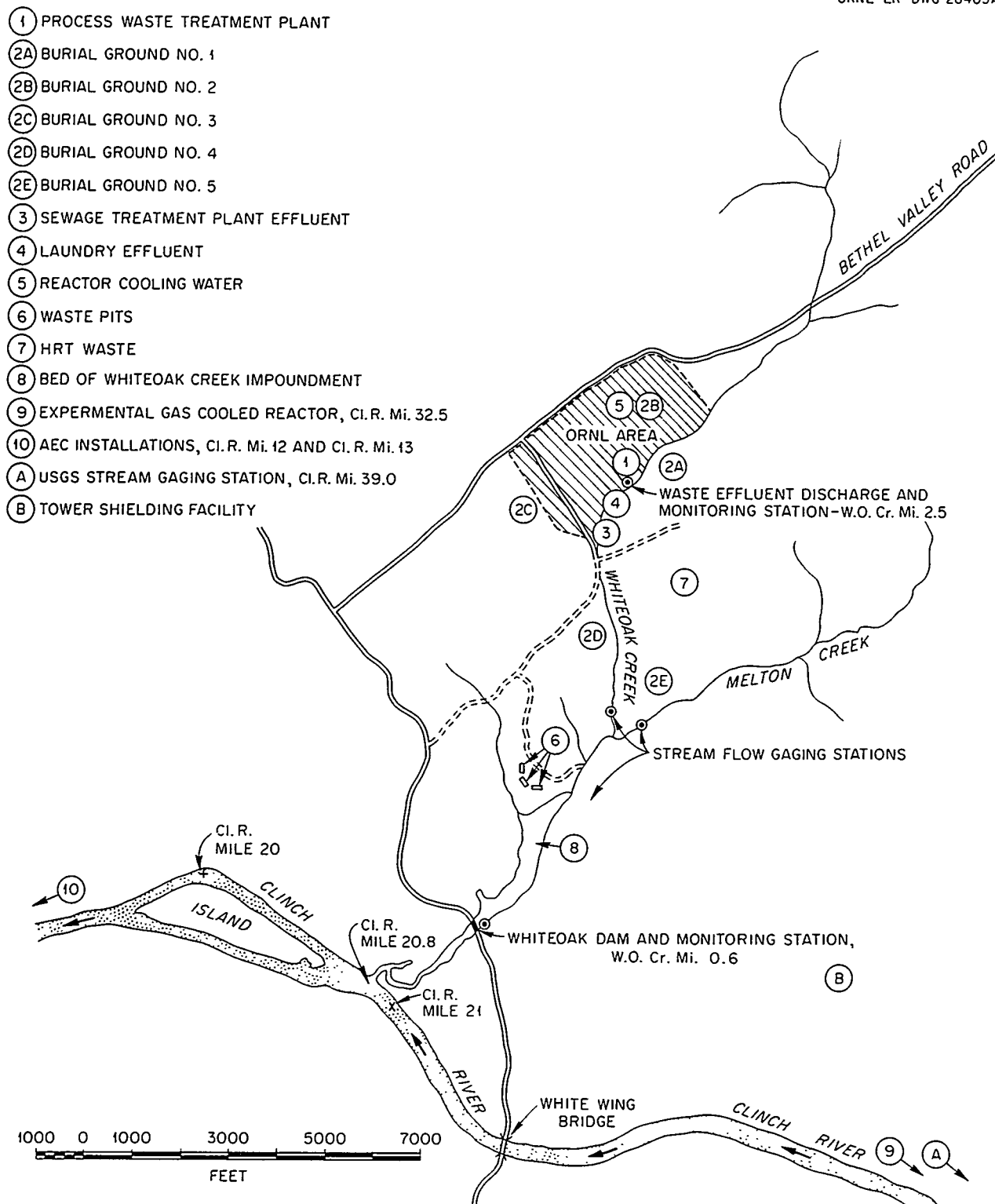


Fig. 4 Sources of Radioactive Contamination in the ORNL Environment

TABLE VI (Continued)

(B) Sources located outside the plant area

Waste pit No. 1
Waste pits No. 2, 3, 4*
Waste pit No. 5 (not yet in use)
Burial grounds No. 4 and No. 5
HRT
Bed of White Oak Lake and White Oak Creek

*Principal Contributors.

In addition to the sources listed above, surface water runoff and fallout can also be included as sources of contamination. Information is available concerning the discharge from the process waste plant, the settling basin, and waste pits 2, 3, and 4. The contributions from the HRT and from the reactor ponds have also been measured and are negligible compared to the other sources. Little information is available concerning the other sources. The available data are summarized in Table VII through XI.

For our purpose it is reasonable to suppose that the contributions from the storm sewers, the sewage disposal plant, the laundry, the abandoned waste pit No. 1, the burial grounds, and from runoff and fallout are normally small compared to the three remaining sources. Table XI gives a comparison of the known amounts of activity added annually to White Oak Creek with the measured amounts entering the river over White Oak Dam. In this table the contributions from the settling basin and from the process waste system have been combined since they enter the creek at the same point. It can be seen from a perusal of this table that with the exception of ruthenium the annual rate at which activity has entered the Clinch River since 1955 has exceeded the

measured rate of discharge to the creek. It is interesting to note that White Oak Lake was drained in October, 1955 so that some connection between that event and the excess activity entering the Clinch River might be inferred.

At present it is not possible to account for this excess activity from any of the known large sources. A recent survey of the White Oak Lake Bed indicates that it contains of the order of 15 curies of Sr^{90} , which is much less than enough to account for the annual excess of 60 curies. Moreover, there is positive evidence to indicate that this material is not coming from the waste pits. There remain only two obvious explanations: (1) The sampling and analytical techniques are in error so that the excess is synthetic, or (2) The excess is real and is coming from a source or sources unknown. In the absence of information to the contrary, the latter explanation will be adopted. It should be understood that this assumption is made by default and that it is subject to modification whenever more complete information becomes available.

Since 1957, nearly all of the ruthenium entering the creek has originated in the waste pits. There is little, if any, evidence to show that any isotope other than ruthenium finds its way from the waste pits to the creek. Moreover, during this same period the fraction of the ruthenium reaching the creek which also reaches the river has retained the nearly constant value 0.3.

Examination of the last three columns of Table XI shows that, since 1953 with the exception of 1958 when a leak of unknown magnitude and duration occurred in one of the waste lines, the strontium excess has ranged quite closely around 60 curies.

TABLE VII (2)

Yearly Volumes and Radioactivity of Discharges of Liquid Wastes to White Oak Creek at ORNL 1953 - 1959^a from the Process Waste System and Settling Basin

Year	Total Volume 10 ⁶ gal	GROSS BETA ACTIVITY		Per Cent of Gross Beta Activity Identified with Specific Radionuclides									
		Settling Basin & Retention Pond Total	Curies	TRE (-Ce) ^b	Ce	Ru	Zr	Ca	I	Sr	Nb	Ba	Co
1953	239.4	429		53.1	2.6	0.8	2.6	8.5	0.2	27.6	1.0	0.8	---
1954	164.3	254		34.6	19.1	0.5	1.0	20.3	0.5	23.3	0.4	0.3	---
1955	210.6	267		30.3	14.7	3.1	0.6	31.6	0.2	18.7	0.6	0.2	---
1956	260.7	273		24.4	12.3	2.0	0.5	42.1	0.1	15.1	1.0	0.2	2.2
1957	272.3	189		25.8	4.4	1.0	10.2	36.5	0.0	18.0	0.7	0.0	1.8
1958	232.0	92		24.0	4.6	1.4	0.7	50.2	0.6	15.3	1.1	0.6	1.5
1959	470.5	181		25.0	16.9	31.7	0.9	11.7	0.1	Sr ⁸⁹	0.5	0.1	2.4
										9.9			
										Sr ⁹⁰			
										0.8			

^aVolume of settling basin effluent measured in weir box with liquid level float recorder. For determination of radioactivity, continuous proportional samples of the effluent are composited for daily gross beta measurements and monthly radiochemical separation and analyses.

^bTrivalent rare earths exclusive of cerium.

TABLE VIII⁽²⁾

Ruthenium Release from Waste Pits
to White Oak Creek

Date	Total Curies	Gallons of Seepage x 10 ⁶
1957	200	4.92
1958	160	4.84
1959*		
Stream at Well 95	30	
Stream at Well 85		
1/59 to 10/59	200	
10/59 to 11/59	60	
11/59 to 12/59	190	
12/59 to 1/60	840	
	1290	1320

* ⁶⁰Co present in streams during 1959; contribute less than 2%
to the total curies reported as ruthenium.

TABLE IX⁽²⁾

Wastes Released from HRT

Release Period Covered	Beta Curies Added to HRT Pond	Beta Curies Released from Pond to Melton Branch	Volume of Release-Gal
Oct. 9 - Nov. 11, 1958	7	2.75	571,000
Nov. 12, 1958 - Jan. 31, 1959		1.65	505,000
Feb. 1, 1959 - Mar. 1, 1959	331	1.1	566,000
Mar. 2, 1959 - July 1, 1959	60	3.1	1,534,000
July 2, 1959 - Oct. 1, 1959	100	1.3	845,000

Principal Contaminants

Ce¹⁴¹⁻¹⁴⁴

Ru¹⁰³⁻¹⁰⁶

Zr - Nb⁹⁵

I¹³¹⁻¹³³

Ba - La¹⁴⁰

Cs¹³⁷

Xe¹³¹⁻¹³³

TABLE X⁽²⁾

Reactor Cooling Water

Demineralizer water from reactor pools is discharged to holding ponds. Total capacity is about 26,000 gallons. Water is discharged about twice monthly from these ponds, generally on week-ends after a decay period such that gamma count rate is less than 100 c/min. Usual count rate is 50 - 90 c/m/ml.

Well counter calibration - 1 c/mr = 1.19×10^{-6} uc/cc

Principal activity is Na²⁴

$$10^2 \text{ c/m/ml} \times 1.19 \times 10^{-6} \text{ uc/c/m} \times 2.6 \times 10^4 \text{ gal} \times 3.785 \times 10^3 \text{ ml/gal} \\ 11.7 \times 10^3 \text{ uc} = 11.7 \times 10^{-3} \text{ curies} = .0117 \text{ curies}$$

TABLE XI

Comparison of Activity Discharged to White Oak Creek
to That Discharged to the Clinch River

		Total	Ce	Curies Ru	Co	Sr	Excess Sr Ce Cs		
1959	Process Waste	181	31	57	21	19			
	Waste Pits	1320		1320					
	Total	1501	31	1377	21	19			
	River	937	92	441	56	77			
	Ratio	0.62	3.0	0.32	2.67	4.05	58	61	31
1958*	Process Waste	92	4	1	46	14			
	Waste Pits	160		160					
	Total	252	4	161	46	14			
	River	544	30	42	55	147			
	Ratio	2.16	7.25	0.26	1.20	10.50	133	36	9
1957	Process Waste	189	8	1	69	34			
	Waste Pits	200		200					
	Total	389	8	201	69	34			
	River	397	13	60	89	83			
	Ratio	1.02	1.63	0.30	1.28	2.44	49	5	20
1956**	Process Waste	273	34	6	115	41			
	River	582	59	29	172	104			
	Ratio	2.13	1.74	4.83	1.49	2.53	63	25	57
1955	Process Waste	267	39	8	84	50			
	River	437	85	31	63	92			
	Ratio	1.63	2.17	3.87	0.75	1.84	42	46	-11
1954	Process Waste	254	49	1	52	59			
	River	384	24	11	22	135			
	Ratio	1.51	0.50	11.0	0.42	2.28	76	-25	-30
1953	Process Waste	429	11	3	37	65			
	River	304	7	26	6	135			
	Ratio	0.71	0.64	8.7	0.16	1.14	70	-3	-31

* During 1958 an unknown quantity of mixed fission products were processed through the sewage disposal plant and thus did not appear as an addition to the Creek.

**Records on the Ru¹⁰⁶ discharge from the waste pits are not available prior to 1957. However, inventory records for 1956 suggest that the value for that year was of the order of 100 curies.

1953-1957 Specific radiochemical analyses were not performed on the wastes discharged from the retention pond to White Oak Creek; therefore, the strontium excess may be accounted for, in part, by this unidentified mixture of fission products from the pond.

The excess of the other two fission products Ce^{144} and Cs^{137} has varied more widely but when positive, has averaged about 36 and 30 curies, respectively. Comparable information is not available for the case of the α emitters; however, the average annual discharge, as seen from Table II, is so small that any contribution from the unknown source will be neglected.

Utilizing the forgoing assumptions, it is now possible to modify the relation (4) to give an estimate of the result in the river of given yearly discharges of isotopes at the major sources. Specifically, it will be assumed that 60 curies of Sr^{90} and 50 curies each of Ce^{144} and Cs^{137} are contributed by the unknown source. In addition, only 30 per cent of the Ru^{106} entering the creek from the waste pits reaches the river. It is further assumed that 100 per cent of the contributions from the waste disposal system and the settling basin reaches the river but that only Ru^{106} is contributed from the waste pits. We obtain, therefore, for the ratio of the concentration in the river to MPC

$$0.172 + 2.8 \times 10^{-3} \left[D_{Sr} + 0.008(0.3D_{Ru} + D_{Ce}) + 0.025(D_{Cs} + D) \right] = X \quad (5)$$

where the D's are the discharge rates in curies per year from all sources except the unknown source in curies per year. It is interesting to note that, if all the D's are zero, i.e., if only the excess is considered, we obtain $X = 0.172$. In all of these computation it should be noted that strontium accounts for over 90 per cent of the value of X.

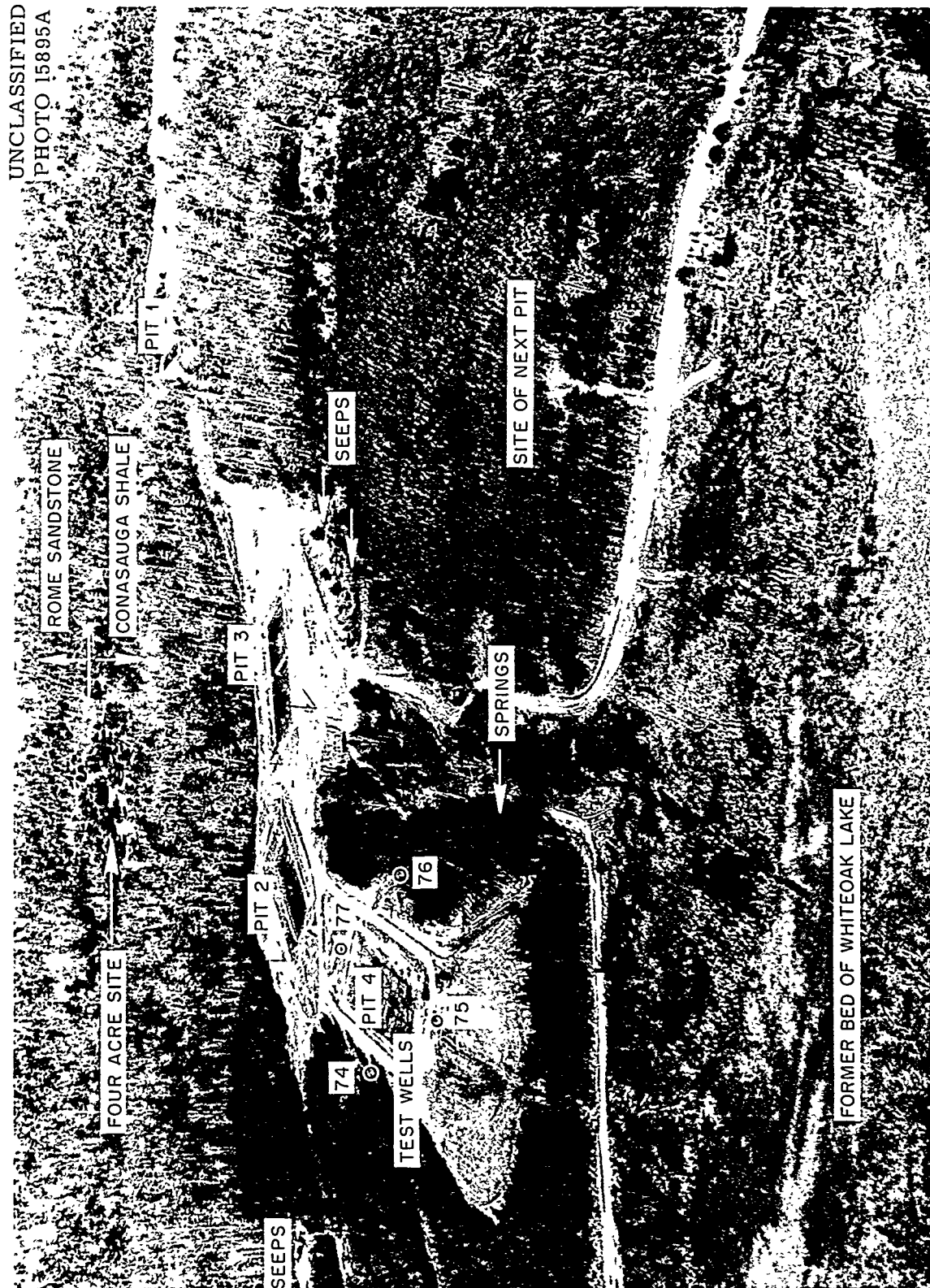
THE EFFECT OF THE WASTE PITS - NORMAL OPERATION

The assumptions and approximations developed in the previous sections may now be used to examine the effect of the waste pit operation in relation to the entire disposal system.

The three pits in current use have been described in detail elsewhere.⁽⁵⁾ Each pit is roughly 200 feet long, 100 feet wide, 15 feet deep and has a nominal capacity at the 12-foot stage of one million gallons. A photograph of the pit area is given in Figure 5. These pits are useful, not only because of their storage capacity but also because of the ability of the soil around them to trap and hold the radioactive constituents in the liquid waste as the liquid waste seeps from them.

Experience to date leads to the conclusion that, with the exception of the element ruthenium, the pit system has been virtually 100 per cent efficient in removing radioisotopes from the liquid which eventually reaches the creek. Recently there has been some evidence of trace amounts of Sr^{90} in the water reaching the creek from the pits; however, the evidence is not conclusive. For the purpose of this analysis it will be assumed that ruthenium is the only radionuclide released. The validity of this assumption has been discussed by Struxness in CF 60-5-29.

Cowser⁽⁶⁾ has presented arguments from which it can be inferred that approximately 7 per cent of the Ru which is contained in the liquid discharged to the pits eventually reaches the small streams en route to the creek. Remembering the strontium ratio for Ru^{106} of 0.008 and the fact that only 30 per cent of the ruthenium which reaches these small streams also reaches the



Aerial View of Waste Pits.

FIGURE 5

river, it follows from equation (5) that a total of 2.1 million curies of Ru^{106} could be handled annually through the pits without exceeding MPC provided that no other radioisotopes were present from any source whatever. This would correspond to an annual discharge to the river of 44,600 curies in agreement with the total indicated in Figure 4. It is instructive to compare the ratio of concentration to MPC contributed by ruthenium to that contributed by other sources. Table XII below gives such a comparison based upon Table XI and equation 4.

TABLE XII

Contributions to the Ratio of Concentration to MPC

<u>Year</u>	<u>From Ru</u>	<u>From Sr</u>	<u>All Others</u>	<u>Total</u>
1959	0.014	0.312	0.009	0.335
1958	0.001	0.365	0.004	0.370
1957	0.001	0.186	0.005	0.192
1956	0.001	0.283	0.028	0.312

From this table it can be seen that in the worst case the Ru has contributed only 1.4 per cent to MPC and only 4.2 per cent of the ratio. On the other hand, the contribution of Sr^{90} is always over 90 per cent of the ratio. It appears, therefore, that the contribution of the waste pits to the total effective contamination of the river has been negligible.

If it is assumed that the quantities of radioisotopes, other than Ru^{106} , remain the same as was reported in 1959, then the quantity of Ru^{106} which could be discharged to the river without exceeding MPC is computed

to be 30,300 curies per year, which corresponds to a total of 1.4 million curies handled through the pits.

The efficiency of removal of radioisotopes in the pit system, apparently 93 per cent for Ru^{106} and 100 per cent for the other isotopes, compares quite favorably with the percentage removal rates in the waste treatment plant. These efficiencies are listed in Table XIII for the period September, 1957 through March, 1959.

On the basis of this comparison plus the evidence given in Table XI, it would appear that the waste pits are by far the most efficient means of removing harmful radioactive material from aqueous waste prior to its discharge to the river.

Once a value for X, the ratio of concentration to MPC, has been chosen as the maximum operating level, it is only necessary to restrict the discharge of activity to the pits in such a way that 2.1 per cent of the ruthenium in that discharge will not increase the total above the chosen value of X. It seems clear that, if the 2.1 per cent value continues to be valid, there is little cause for concern until the discharge to the pits reaches the neighborhood of 10^5 curies annually. On the contrary, if one considers the fact that in 1959 the pits received 24,700 curies of Sr^{90} and if that material had been processed through the waste treatment system rather than having been sent to the pits, the result would have been an increase of over 4000 curies in the Sr^{90} discharged to the creek in that year because of the average efficiency of only 84 per cent removal applicable to this system.

TABLE XIII (7)

Removal of Radionuclides by Treatment
Monthly Composite Samples

	Percentage Removal						Curies Removed					
	Gross Beta Activity	Sr ⁸⁹⁺⁹⁰	TRE*	Cs ¹³⁷	Co ⁶⁰	Ru ¹⁰⁶	Sr ⁸⁹⁺⁹⁰	TRE*	Cs ¹³⁷	Co ⁶⁰	Ru ¹⁰⁶	
1957												
September	67	87	89	9.5	51	7.6	3.7	2.6	0.62	0.02	0.19	
October	68	73	77	19	23	0	2.6	2.7	0.84	0.12	0	
November	87	80	80	21	72	83	2.4	1.9	0.23	0.12	0.74	
December	79	84	87	40	83	67	2.4	2.6	0.74	0.21	0.31	
1958												
January	53	74	74		76	65	3.6	2.7		0.19	0.38	
February	57	66	67	42	56	42	2.3	2.4	1.4	0.17	0.18	
March	73	85	88	32	69	66	4.7	5.1	1.1	0.16	0.30	
April	70	84	86	40	74	74	5.3	5.5	1.6	0.25	0.26	
May	59	78	80	0	61	69	6.0	5.9	0	0.34	0.36	
June	57	84	86	0	67	24	5.2	5.2	0	0.25	0.09	
July	55	85	86	0	70	30	10	7.6	0	0.30	0.06	
August	54	85	89	2.1	64	73	7.4	7.5	0.31	0.20	0.70	
September	77	79	81	48	73	38	9.6	9.8	8.2	0.82	0.20	
October	84	89	71	80	82	84	6.5	7.8	11	0.50	0.12	
November**		82	85	84			5.0		8.5			
December	84	90	89	84	65	59	7.3	8.2	12	0.54	0.12	
1959												
January	86	87	89	86	78	47	7.8	12	8.1	4.6	0.33	
February	83	79	86	84	78	73	5.4	8.9	7.0	1.3	0.46	
March	90	88	95	84	65	72	7.0	26	4.8	0.78	1.1	
						Total	104.0	124.0	66.0	10.9	5.9	

The fact is that the contribution of Ru^{106} from the waste pits has been and continues to be completely negligible, not only compared to the Sr^{90} which reaches the creek from other sources but on an absolute basis as well.

HAZARDS ASSOCIATED WITH THE WASTE PITS

The optimistic assessment of the waste pit operation presented here is predicated upon the assumption that the pits will continue to function as they have in the past and that the complete removal of radioisotopes, other than Ru, by these pits is, as the evidence strongly indicates, indeed the case. There are, however, conceivable but not necessarily credible circumstances under which the pits could release strontium, cesium, and cerium in quantities greater than those heretofore recorded.

The results of the release of the liquid waste directly to the river will be considered first. The magnitude of the associated hazard will, of course, depend upon the quantity and nature of the activity present in the pits at the time of the release. It will also depend in a complex way upon the flow in the river at that time. Since the pits are operated at constant volume, the addition and loss rates of the liquid are about the same; namely, 10,000 gallons per day on the average. The total volume usually present in the pits is about two million gallons. The quantities of the important nuclides, based upon the yearly discharges from 1956 through 1959 which were present in the pits at the end of 1959, are listed in Table XIV.

Upon using the strontium ratios listed in Table III it can be seen that the total activity in the liquid in the pits was equivalent to 9,500 curies of Sr^{90} of which 88.4 per cent was due to the presence of strontium

Waste Pit System Inventory 1959 Year End

Isotopes	Total	Curies Liquid in pits	Ground Inventory
Ru ¹⁰⁶	195,000	46,000	149,000
Sr ⁹⁰	37,000	8,400	28,600
Ce ¹⁴⁴	26,000	6,000	20,000
Cs ¹³⁷	150,000	34,000	116,000

and only 11.6 per cent to the presence of other isotopes. Ruthenium accounted for only 3.9 per cent of the total hazard.

Quite clearly the hazard associated with this fission product inventory is due almost entirely to the presence of Sr^{90} . The discharge of this material to the river over a short period of time would result in an increase in the concentration to a value many times the MPC although this value would not persist for a very long period of time.

Let us consider the means whereby the contents of the pits could be suddenly emptied into the river. There are four agencies which seem capable of causing complete or partial rupture of the pits. These are as follows: earthquakes, floods, earthslides caused by heavy rains, and sabotage. Because of the location of the pits and the geological history of the area, it is not credible that either of the first two agencies could initiate a failure. The East Tennessee Region is in a relatively quiescent, seismic area; and the occurrence of a destructive shock is considered virtually impossible. The waste pits are located above elevation 800, and the maximum flood level is 777.

Probably the most likely natural cause of rupture would be heavy rainfall sufficient to initiate an earth slide on the east side of waste pit No. 4. This could result in the release of all the waste in pit No. 4 to the river. Normally this pit contains much less activity than the other two; however, at worst it would contribute one third of the total stored activity. This event is considered credible but unlikely. Moreover, it is proposed to abandon pit No. 4 in the near future.

Finally, well executed sabotage through the use of sufficient explosives to blow large holes in the pit walls could result in the discharge of the contents to the Clinch River. This contingency is guarded against by the normal plant security procedures. It is perhaps true that the present procedures are not adequate to prevent sabotage of these pits.

One other natural phenomenon which could result in widespread damage must be considered. This is the passage of a tornado across the pits. The result would be the removal of the liquid in the pits and its subsequent deposition over a wide area along the path of the storm. It is possible to postulate a catastrophic sequence of events following such an occurrence. This accident is not considered credible. In the first place the incidence of tornadoes in the Oak Ridge area is quite small although one did pass through a part of the reservation in 1951. In addition to the low frequency it would be necessary that the path of the storm bring it into coincidence with the location of the waste pits, an event of very low probability. Moreover, future waste pits are designed in such a way that there will be a minimum of six feet of rock above the surface of the liquid. This should effectively prevent any liquid from being carried away by a storm.

Referring again to Tables III and XIV, it can be seen that at the end of 1959 the equivalent of 32,300 curies of strontium was fixed in the soil near the pits. Of this, 28,600 curies were actually due to strontium, and the remaining 3,700 curies equivalent were due to the other isotopes. This large inventory, representing 3.4 times that stored in the pits, presents a potential hazard only if there exists some mechanism which is

likely to cause it to enter the stream.

The observations made to date⁽⁶⁾ lead to the conclusion that, with the exception of ruthenium, all of the nuclides discharged into the soil from the waste pits remain in the soil very near to the pits. These observations, made by taking samples from wells drilled for the purpose near the pits, have indicated that none of the isotopes other than ruthenium have migrated through the soil. Despite this fact, the possibility exists that the ability of the soil to hold these chemicals may become inhibited for some reason or reasons not yet known. Moreover, it seems probable that at some point the soil will become saturated with the materials which seep from the pits and will then have no further capacity to hold additional quantities. For these reasons it is necessary to constantly monitor the area near the pits in order to detect any movement of the ground inventory toward the stream bed. Should such a trend be detected, it will be necessary to abandon operations in the location where this is taking place. The offending pit or pits would be emptied and sealed. This will result in a lowering of the water table at that point and should prevent any further movement of the ground inventory.

MAXIMUM CREDIBLE ACCIDENT

The maximum credible accident to the pits is considered to be the introduction into the river of the entire contents of the waste pits. The magnitude of the hazard will depend directly upon the activity present in the pits at the time the incident occurred. It will depend in a more complicated

way upon the flow in the river during and following the release. The weekly averaged flows in the river for the past four years are given in Figure 2. The lowest flow recorded in the Clinch River at the mouth of White Oak Creek was 150 CFS and the maximum was 27,700 CFS. It is not entirely clear whether high flows or low flows result in a greater hazard. High flows permit the activity to move downstream more rapidly and result in greater dilution; however, less time is available for emergency action to be taken, and less reduction in activity due to dispersion occurs. Moreover, under some conditions high flow will result in Clinch River water entering the Harriman water treatment plant on the Emory River. Low flows, on the other hand, will result in higher concentrations and a longer duration of high concentration at any one point. This is offset to some extent by greater dispersion and better mixing.

In order to get an estimate of the results of this accident, it will be assumed that the waste pits contain two million gallons of liquid. It appears that a concentration of $8 \mu\text{c/ml}$ of ruthenium is about the maximum which can be allowed in the pits without exceeding the limits subsequently set for normal discharge to the river.⁽⁶⁾ Thus, if the other constituents are present in the same ratio as that given in Table XIV, then the quantities of the various activities present are as follows:

TABLE XV

Assumed Waste Pit Inventory For Maximum Credible Accident

Ru ¹⁰⁶	60,600 curies
Cs ¹³⁷	44,800 curies
Sr ⁹⁰	11,000 curies
Ce ¹⁴⁴	7,900 curies

The strontium equivalent of this mixture is 12,500 curies.

The river flow at the mouth of White Oak Creek at the time of the accident will be taken to be 5,000 CFS. No loss of activity by dispersion, decay, or other means will be assumed; and dilution will be taken as the only method by which the concentration is reduced. Thus, if the time over which the discharge occurs is T seconds, then the concentration in the river at mile $13.4(K - 25)$ will be

$$12,500 / (T \cdot 141.6) = 88.3 / T \text{ } \mu\text{c/ml}$$

In this approximation the duration of this concentration at a point downstream will be just the time T , so that the total dose from the activity will be $88.3 \text{ } \mu\text{c sec/ml}$. Now the occupational MPC for the mixture is $8 \times 10^{-7} \text{ } \mu\text{c/ml}$ so that the exposure resulting from this slug of activity is on the average roughly equivalent to that received from $88.3 / (8 \times 10^{-7} \times 3.15 \times 10^7) = 3.5$ years of exposure to continuous occupational tolerance.*

Actually the slug of activity diffuses as it goes down river, and the time required for the slug to pass a given point is much longer than just the duration of the release. Estimates made by the staff of TVA⁽⁸⁾ indicate that this time may be roughly approximated by the formula

$$T_s = T_{\text{transp}} + 1.5 T$$

where T_s is the time required for the slug to pass a given point, T_{transp}

*It is appreciated that a short-term high level exposure may be more serious than a long-term low level exposure; however, this comparison should give a reasonable estimate of the order of magnitude of the hazard.

is the average time for water to reach the point in question from the site of the incident and T is the duration of the incident. Thus, for releases of short duration the time required for the activity to pass a given point is roughly equal to the transport time between the point of release and the point in question.

Obviously the concentration is actually less than that calculated in the previous approximation; however, the time integral is about the same. The durations of the incident for various locations on the Clinch are given in Table XVI.

Because of the relatively long times involved, it would appear feasible to give warning downstream in time to prevent any dangerous overexposures.

Dilution alone in the Tennessee River would account for reduction of at least six in the concentration of activity by the time it reached Chattanooga. The total dose delivered, however, would occur over a period of about 12 days. Thus, completely neglecting dispersion in Watts Bar Reservoir, the dose received at Chattanooga would be equivalent to about 0.6 years at occupational tolerance levels. Dispersion in the Reservoir would probably reduce this by at least a factor of ten.

CONCLUSIONS

The following conclusions may be drawn from this analysis:

- (1) The disposal of liquid waste in the open pits at ORNL has contributed an insignificant amount of the total activity hazard released to the environment from ORNL. Provided that the pits continue to behave as they have in the past and provided the amount of activity stored in the

TABLE XVI

Time Required for Contamination from a Short
Duration Release at White Oak Creek to
Pass a Point

River Flow	Mile 13.4 (K-25)	Mouth
2500 CFS	1.3 days	8 days
5000 CFS	.75 days	3.7 days
7500 CFS	.45 days	2.5 days

pits is maintained at a level low enough so that the maximum discharge rates to the river are not exceeded, this method of disposal can be considered quite satisfactory.

(2) The maximum credible accident is serious and could be quite costly, but it is not catastrophic. By this is meant that no widespread injury or loss of life is anticipated but that the cost in lost time and property damage could be quite high. The high levels of activity to be expected following the accident could cause widespread damage if the contaminated water was allowed to enter municipal water treatment facilities and industrial plants. The establishment of a warning system to prevent the intake of contaminated water and of survey procedures to determine the extent of the hazard would considerably reduce the damage. Despite this the resultant long-term low level contamination of Watts Bar and Chickamauga Reservoirs would result in an extremely undersirable situation.

(3) The primary hazard is due to the presence of large quantities of Sr^{90} in the pits. The use of the pits for the disposal of materials other than Sr^{90} is almost two orders of magnitude less hazardous.

GENERAL RECOMMENDATIONS

The following recommendations are made:

(1) A system of monitoring all sources of contaminated material which can reach the river should be installed and put into operation just as soon as possible. Investigations of the sampling and analytical procedures used to determine the rate of discharge of activity to the river

should be instituted at once and the source of the "excess" activity determined. These studies should be accompanied by the acquisition of sufficient data concerning the concentration of activity in the river so that the result of various discharges can be predicted with certainty.

(2) A procedure for warning downstream communities in case of the release of a dangerous amount of activity should be developed and put into operation at once. Procedures to monitor the course of the released activity, to determine the hazards involved, and to make pertinent information available to the proper authorities should be included. The appropriate courses of action including water intake shutdown and evacuation of persons living on or near the river should be worked out in detail.

(3) The development of methods to reduce the amount of Sr^{90} present in the wastes stored in the pits would greatly reduce the hazard.

(4) The program directed at the study of conditions under which fission products move through, or adhere to, the soil should be intensified in order to better understand the way in which ground disposal can be safely used. In connection with this study it is desirable to re-examine the methods currently in use in order to make sure that any movement of fission products toward the stream will be detected at an early stage.

(5) Since it is not possible to determine a priori the characteristics of a given pit with respect to the retention of radionuclides, all new pits should be tested with low level wastes to insure that they operate in a satisfactory fashion before any large amount of active material is discharged to them.

(6) Steps should be taken to improve the security of the pits from tampering or deliberate sabotage.

RECOMMENDATIONS WITH RESPECT TO OPERATION

As has been pointed out previously, it is assumed that, once a maximum permissible average concentration of radioactive contaminants in the Clinch River has been chosen, it is required to operate the waste disposal complex in such a way that this average is not exceeded. The value chosen is the non-occupational MPC set forth in BS Handbook 69. However, in order to take care of contingencies and the fact that other operations may also contribute to the contamination, 30 per cent of this value will be chosen as the operating standard for ORNL waste disposal operations.

Thus, as a first operating criterion, it is recommended that the amount of activity discharged to the river shall be such that the average value of X , as defined in the equations developed in Section 3, shall not exceed 0.3 averaged over any twelve-month period.

Now the rate at which activity is discharged to the river is affected not only by the amount of activity processed through the various disposal agencies but also by climatic conditions. Thus, it is not possible to discharge activity to the river at a uniform rate. For this reason a second criterion is proposed; namely, that the discharge rate shall be such that the average value of X for any seven-day period shall not exceed 3.0.

Clearly, the contribution permitted from the waste pits will depend upon the extent to which the foregoing limits are approached by other sources of activity. Neither the waste pits nor the settling basin are

susceptible to rapid control. Process waste water, on the other hand, can be impounded to some extent in the equalization basins. Thus, it appears that the total amount of activity permitted in the waste pits should be maintained at a level which when related to the expected activity from the settling basin cannot result in the excess of released activity over the limits suggested. This upper limit to the total activity also effects the magnitude of the maximum credible accident. In the illustration discussed the upper limit was set on the basis of $8\mu\text{c/ml}$ of ruthenium in the pits. In the past the concentration has exceeded this and at times exceeded $25\mu\text{c/ml}$. Clearly this situation must be rectified.

In order for any control system to be effective, it is absolutely necessary that the discharge of waste be regulated. This may mean that at times those processes which generate waste may be forced to shut down for varying periods of time so that the average concentrations may be maintained. This is a penalty which must be accepted until more positive means of decontaminating, storing, and metering radioactive waste are developed.

REFERENCES

1. Maximum Permissible Body Burdens and Maximum Permissible Concentrations of Radionuclides in Air and Water for Occupational Exposure, U. S. Department of Commerce, Bureau of Standards Handbook 69.
2. ORNL Health Physics Division records.
3. Private communication, K. E. Cowser.
4. Private communication, H. H. Abee.
5. K. E. Cowser and F. L. Parker, Health Physics, Vol 1, 152-163 (1958).
6. K. E. Cowser, Potential Hazard of Ruthenium in the ORNL Waste Pit System, ORNL-60-3-93 (March 11, 1960).
7. "Health Physics Division Annual Progress Report for Period Ending July 31, 1959," ORNL 2806.
8. Private communication, E. G. Struxness.